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Regarding the Frequency-Based Equation of State of Segletes

by Steven B. Segletes

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Abstract

Clarifications and observations are made regarding Segletes' equation of state, in response to recent comments and criticisms. A theoretical justification for the form of the equation of state is laid out in more precise detail. The model is shown to produce high-quality fits to shock-, as well as cold-compression data, especially when accounting for the experimental variability of the model input parameters. The falseness of the correspondence between the volumetric and vibrational spring constants for an atomic lattice, previously inferred from Segletes' equation of state, is proven by alternate means. The non-Grüneisen behavior of aluminum is addressed, and a brief note is made regarding the nature of isentropic transitions.

Acknowledgments

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1. Background

Since the introduction of “An Equation of State for Metals [1],” a good deal of interest has been shown in the work and its sequel papers [2–4]. Some of that interest has come in the form of questions from would-be users of the model. Other helpful comments and suggestions came from readers interested in the subject, and, not without precedent, criticisms were leveled by various reviewers. Hence, in this report, further explanation is provided to buttress the theoretical basis for some of the model’s features. This paper answers reviewer’s questions, points out some subtler features, and ties up loose ends regarding Segletes’ equation of state for metals.

The earlier documents [1–4] should be consulted for full understanding of the model. To facilitate a proper frame of mind for the current reading, however, a cursory review of the work leading up to this report is given here. Segletes [1] introduced an equation of state for metals, which has the unique distinction of being both a frequency-based (*i.e.*, characteristic-temperature-based) approach, as well as a model that incorporates our current understanding of the lattice potential.

Earlier frequency-based approaches, like those of Slater [5], Dugdale and MacDonald [6], and Vashchenko and Zubarev [7] did not have lattice-potential data available to them to critically test their models. Later models, like those of Rose *et al.* [8], Vinet *et al.* [9, 10], Baonza, Cáceres, and Núñez [11], and Baonza *et al.* [12], though reflecting a more current understanding and recent data of the lattice energy potential, remain, by contrast, in the domain of density and interatomic distance, rather than that of the lattice vibrational frequency spectrum. One point that this report shows is that the characteristic temperature of the lattice, which relates to the vibrational frequency spectrum, is necessarily the natural variable in an equation of state for solids, in preference to, for example, interatomic distance.

Segletes [1] gave his equation as

$$p \psi - E = E_b \left\{ \left[\left(\Theta / \Theta_0 \right)^K - 1 \right] + K (K - 1) \left(\Theta / \Theta_0 \right)^K \ln(\Theta / \Theta_0) \right\} . \quad (1)$$

In this equation, Θ is the characteristic temperature of the lattice, which is (according to Grüneisen theory) taken as a function of volume only. The variables p and E are the pressure and specific internal energy respectively; K is a parameter, given by $C_0/(\Gamma_0 E_b^{1/2}) = \eta/(3 \Gamma_0)$, where C_0 is the reference bulk sound speed at zero temperature and pressure, E_b is the specific lattice binding energy, and η is the lattice anharmonicity parameter given by Rose *et al.* [8]; and $\psi = V/\Gamma = (\partial E/\partial p)_V$ is a thermodynamic variable introduced [13, 14] for ease in manipulating the governing equations. When this variable, ψ , is likewise retained as a function of volume only (*i.e.*, independent of temperature) the relationship that relates the characteristic temperature to the Grüneisen function becomes

$$\frac{\Theta'}{\Theta} = -\frac{1}{\psi} , \quad (2)$$

where the prime denotes differentiation with respect to specific volume, V . The generalized cold curve associated with eqn (1) is

$$E_c = E_b \{1 - (\Theta/\Theta_0)^K [1 - K \ln(\Theta/\Theta_0)]\} \quad (3)$$

and

$$p_c = \frac{E_b K^2}{\psi} (\Theta/\Theta_0)^K \ln(\Theta/\Theta_0) . \quad (4)$$

In the original paper [1], in which eqns (1)–(4) were introduced, a linear form for $\psi(V)$ was adopted, based on previous work on the thermodynamic stability of the Grüneisen equation of state [13–15]. Excellent fits to cold-compression data prevailed for the nine metals studied, with the resulting constrained predictions of shock-compression behavior also being exceptional into the megabar regime (with one explainable exception). On the lattice expansion side of equilibrium, an adequate correlation was achieved to the universal lattice potential of Rose *et al.* [8]. Eqn (3), which was the actual genesis of Segletes' model, was inspired by the universal potential of Rose *et al.* [8]. This inspiration, by which a functional correlation was drawn

between the characteristic temperature and Rose's lattice parameter term, was not addressed in the greatest detail. This report, thus, lays out a much firmer justification for the functional form of Segletes' model than was, perhaps, implied in his original work [1].

In the next papers to have appeared on the subject, Segletes and Walters [2, 3] examined the use of an empirical power-law fit for $\psi(V)$, in preference to the linear fit used in the original work [1]. The fit to compression data remained exceptional, while at the same time, an excellent correlation was achieved in the expansion end of the lattice energy potential by relating, in a linear fashion, the lattice anharmonicity parameter, η , of Rose *et al.* [8] to the power-law exponent on ψ . By ignoring higher order terms, the power-law $\psi(V)$ could be made, to reproduce the historical Grüneisen models of Slater [5], Dugdale and MacDonald [6], as well as Vashchenko and Zubarev [7] merely through appropriate selection of a single parameter—the power-law exponent on $\psi(V)$. By retaining the higher order terms, however, important differences arose between the older and current models.

In a more recent paper, Segletes [4] took the idealized quasi-harmonic form of his model (defined when the K parameter takes on a value identically equal to unity and the Grüneisen function varies as $\Gamma \sim V^{1/3}$ [*i.e.*, $\psi \sim V^{2/3}$]) and was able to show how this form reduces to the harmonic approximation for an atomic lattice as the Grüneisen function approaches zero. Such a result is important, since the harmonic approximation is the viewpoint historically used to derive many of the results relating to the behavior of atomic lattices.

Furthermore, Segletes [4] inferred from his model the falseness of the assertion that the volumetric spring constant, $dF/d\lambda$ (where F is the interatomic force, and λ is the lattice spacing) is, in general, the same spring constant that drives the vibrational frequency of the lattice (termed as $\partial F_{vib}/\partial x$). The assertion of a proportionality between the volumetric and vibrational spring constants derives from one-dimensional (1-D) lattice theory in which the motion of an atom is influenced only by interactions with its nearest neighbors. In such an idealized configuration, the spring constants $dF/d\lambda$ and $\partial F_{vib}/\partial x$ are indeed equal to each other. That such a proportionality of spring constants would hold, in the general case of real atomic lattices, is an assumption that has pervaded the literature on the subject, having been used, in part or whole, by Slater [5], Dugdale and MacDonald [6], Brillouin [16] and, more recently, Guinea *et al.* [17].

In this report, a simple example is offered that contradicts the assumed correlation of $dF/d\lambda$ and $\partial F_{vib}/\partial x$, thereby disproving the basis for such an assumption. Such a refutation of proportionality is vital in promoting the acceptance of Segletes' model, since the model was shown [4] to deny such a proportionality. A qualitative explanation is offered as to why there should arise a functional discrepancy between $dF/d\lambda$ and $\partial F_{vib}/\partial x$.

2. Justification for the Form of Segletes

Segletes' model [1] was not derived from first principles of physics and might, therefore, seem to call into doubt the scientific basis of his form. On the contrary, the justification arises through the use of mathematical arguments to link the homogeneous solution of the Grüneisen cold curve to a term in the lattice energy potential of Rose *et al.* [8]. Let us proceed to prove this point.

The Grüneisen equation of state may be expressed in a variety of ways, but amounts to the following:

$$p - p_{ref} = (E - E_{ref})/\psi . \quad (5)$$

In this equation, the reference functions are the pressure and specific internal energy states along a known reference curve, such as an isotherm, isentrope, or Hugoniot. Since, for Grüneisen materials, p_{ref} , E_{ref} , and ψ are functions of volume alone, the solution to the Grüneisen equation of state, eqn (5), amounts to determining ψ and $G(V)$, which satisfy

$$p\psi - E = G(V) . \quad (6)$$

Along the cold curve, where $p_c = -E'_c$, one obtains the differential equation

$$\psi E'_c + E_c = -G(V) . \quad (7)$$

A complementary solution to the homogeneous form of eqn (7), corresponding to $G(V) = 0$, is

$$E_c = C\Theta \quad , \quad (8)$$

which may be verified through the use of eqn (2), where C is an integration constant. Incidentally, it is this complementary solution that constitutes the cold curve in both the Einstein and the Debye equations of state (*i.e.*, no particular solutions were offered).

When Rose *et al.* [8] made their case for a universal form of the lattice energy potential, they proposed a form given by

$$E_{pot} = -E_b (1 + a + 0.05 a^3) \exp(-a) \quad , \quad (9)$$

where E_b is the specific lattice binding energy, and a is a dimensionless parameter defining the relative spacing of the lattice atoms, given by

$$a = \eta \left((V/V_0)^{1/3} - 1 \right) \quad . \quad (10)$$

In eqn (10), the parameter η is called the anharmonicity factor. Note that, by lattice energy potential, we refer to the cold energy expressed relative to the infinite lattice-separation condition:

$$E_{pot} = E_c - E_b \quad . \quad (11)$$

The form of Rose *et al.* [8] is such that the potential energy could be expressed in terms of a single independent variable, a , given by eqn (10). To a very good order, this a variable scales the universal potential to that for each material through a single parameter, η . In their case, this independent variable is expressable in terms of specific volume, as in eqn (10), or alternately in terms of lattice spacing, λ , as

$$a = \eta (\lambda/\lambda_0 - 1) \quad . \quad (12)$$

We note here that, though adequate for measures of compression, interatomic distance is not the natural variable in defining the thermal behavior of a lattice, and thus conclude that it is unlikely

that a thermal equation of state, of general form, would arise from the Rose potential, if the independent variable is left in terms of interatomic distance.

If a general thermal equation of state were to exist of Grüneisen variety, expressible in terms of a single independent variable, then we can see that the characteristic temperature, Θ , is the natural choice for that variable, since both it and its spacial derivatives directly tie thermal behavior to compressive behavior. This case is clearly made by observing that Θ relates to thermal behavior of a lattice by way of the Grüneisen function, Γ , according to eqn (2), where, for reminder, ψ equals V/Γ . Simultaneously, eqn (8) shows how $C\Theta$ is a complementary solution of the Grüneisen cold-compression curve (or alternately, the lattice potential).

The thermodynamic variable Θ is unique in this regard. However, there is nothing new in the assertion about the importance of Θ and its derivatives. Slater [5], Dugdale and MacDonald [6] and others have attempted to relate the cold curve to the Grüneisen function (a derivative of Θ). They recognized Γ as being key to a general equation of state but did not have, at the time, the universal potential of Rose *et al.*, or the cold-compression data that support it, at their disposal for comparison.

Segletes [1] did not know, *a priori*, the makeup of the G function that might satisfy a general form of eqn (6), either in terms of $G(V)$ or, to be truly general, in terms of $G(\Theta)$. However, he did know that *if* a single-variable, general, thermal equation of state exists, then it must satisfy or approximate the universal lattice potential of Rose *et al.* [8].

Ignoring, for the moment, the higher order, a^3 correction term of eqn (9) (as have many sequel authors, including Rose himself), the terms of Rose's potential are $\exp(-a)$ and $a \exp(-a)$. Recalling that $E_c = C\Theta$ is a complementary solution to the Grüneisen cold curve, we shall proceed, for the moment, as if this term constitutes the totality of the complementary solution space. Though it is possible, under unique boundary conditions, that the complementary solution to a differential equation does not appear in the particular solution, such a scenerio is highly unlikely in the general case. Proceeding on this basis, we may logically conclude, therefore, that if a general thermal equation of state in one variable exists at all, then Θ is the independent variable and must correspond to a term or term grouping in the potential of Rose *et al.* [8]. The

possibilities here are quite limited, and we see that Θ may take on one of only three possible forms: either $\Theta \sim \exp(-a)$, $\Theta \sim a \exp(-a)$, or Θ goes as the sum of these two terms.

The second possibility is shown immediately not to be plausible, since $\Theta \sim a \exp(-a)$ would have Θ equaling zero at ambient lattice spacing. The third possibility, involving the sum of the two terms, would essentially leave the energy potential, eqn (9), proportional to Θ , which amounts to Debye's or Einstein's equation of state, depending on how Θ were defined. Thus, Segletes [1] proceeded to investigate the first and only remaining case. With $\Theta \sim \exp(-a)$ as the subject of his investigation, a G function needed to be determined so as to be compatible with the universal potential. It was intellectually satisfying to find that taking the absolutely simplest form of G in terms of Θ (i.e., $G = A\Theta - B$) where A and B are constants, then the general solution to the Grüneisen equation of state, eqn (9), is

$$E_c = B - C\Theta(1 - A/C \ln \Theta) . \quad (13)$$

This solution, based on the possibility that $\Theta \sim \exp(-a)$, may be readily converted into the form of Rose *et al.* (minus the a^3 correction), given appropriate selection of constants A , B , and C .

We thus have an equation of state form, in terms of Θ , that can at least be made to fit the universal lattice potential and is compatible with the complementary solution of the Grüneisen cold curve. This form has come with certain assumptions and conditions, so let us review them. First, this form asserts the temperature independence of the Grüneisen function, which seems true for many materials over a wide range of pressure and volume. Second, we accept the form of the interatomic potential put forth by Rose *et al.*, though we will drop the cubic correction term, in the hope that the term will not be necessary, when results are expressed in terms of the natural variable, Θ , rather than λ or V . Third, we assert that the complementary solution to the Grüneisen potential should show up as a term in the particular solution, in this case, corresponding to a term in the potential of Rose *et al.* Such a mapping would be a mathematical certainty, unless an unusual boundary condition governed, in the general case, to zero out the complementary solution from the particular solution. Fourth, a complementary solution to the Grüneisen potential was determined, and is proportional to the characteristic temperature, Θ .

Fifth, the only term in Rose's potential to which this Θ term might correspond and still produce a new and meaningful equation of state, is the $\exp(-a)$ term. This mapping may also be expressed as $a = -\ln(\Theta/\Theta_0)$.

This mapping mathematically corresponds, for eqns (1)–(4), to the idealized $K = 1$ case, which constitutes one part of the definition for a quasi-harmonic material [4]. The other part of the quasi-harmonic definition, that $\Gamma \sim V^{1/3}$, was shown [4], by integration of eqn (2), to imply for the quasi-harmonic case only, that

$$a = -\ln(\Theta/\Theta_0) = 3 \Gamma_0 (\lambda/\lambda_0 - 1) \quad (\text{Quasi-Harmonic}) \quad (14)$$

Were actual materials indeed quasi-harmonic, then this mapping for the a variable provides the exact definition used by Rose *et al.* [8], given in eqn (12), since Segletes [1] showed that the anharmonicity parameter, η , for the $K = 1$ case, exactly equals $3 \Gamma_0$. The conclusion one may draw is that the universal lattice potential of Rose *et al.* [8], minus the cubic correction term, corresponds exactly to the zero-temperature potential associated with the idealized form of Segletes' equation of state—namely, the quasi-harmonic form.

To account for nonidealness of real materials, Rose *et al.* [8] introduced the higher order cubic term to their cold-energy potential. Segletes [1], to accomplish a similar correction for nonideal effects, relaxed the mapping from the idealized form, $a = -\ln(\Theta/\Theta_0)$, to a looser form, $a = -K \ln(\Theta/\Theta_0)$, where the parameter, K , was assumed constant and determinable from known physical properties. Though this relaxation of the mapping was performed [1] *ad hoc*, so as to facilitate a fit to the data, the author will explore, in a subsequent report, how such a relaxed mapping, with the K parameter being allowed to vary slowly, may correspond to other, as yet undetermined, complementary solutions to the Grüneisen potential. The remainder of Segletes' original paper [1] showed how such a mapping, $a = -K \ln(\Theta/\Theta_0)$, onto the potential of Rose *et al.*, could model the thermal, as well as compressive, behavior of a wide variety of metals.

We note that this justification for eqn (3) is not a first-principles derivation from lattice potential theory. Desirable as that would be, the lack of such a derivation does not indicate an

absence of physics in Segletes' model. The physics of the model is that intrinsic to the Grüneisen equation of state and the lattice potential of Rose *et al.* [8], which is supported by ample data. The Grüneisen equation indicates that a general thermal equation of state, if it exists, ought to contain a $C\Theta$ term. The work of Rose *et al.* shows a universal potential in terms of the lattice-spacing variable, a . The contribution of Segletes [1] was in providing a logical link between these existing results, by showing that one of terms in Rose's potential, cast in the variable a , should correspond to the Θ term, as dictated by the governing differential equation.

3. Quality of Fits to Shock Data

In response to the criticism that the fits to shock-compression data, as reported in reference [1], are "merely adequate," the author is forced to refer such readers back to that reference. That paper "intentionally avoid[ed] taking the approach of tuning all available parameters... to avoid the criticism of so-called 'knob turning'." Instead, it took the approach of deriving all quantities solely from the atomic lattice data provided by Rose *et al.* [8] and handbook data for the thermal expansion coefficient, α . Anyone who has tried to obtain published values for the reference value of the Grüneisen function, Γ_0 , for example, will have noted the wide scatter (10% to 50% or more) in published values. Furthermore, if one tries to construct values for Γ_0 using handbook data and the relation $\Gamma_0 = 3\alpha C_0^2/C_V$, where C_V is the specific heat, the scatter is equally wide because of published variations and thermal variability of the component ingredients to Γ_0 .

So, if one wishes to improve the quality of the fits to shock data over that originally reported [1], one simple and wholly justifiable step would be to select values for Γ_0 , based on the published ranges of Γ_0 or, alternately, published values of α , C_0 , and C_V . The results for palladium, for example, reported in Segletes [1], were based on a Γ_0 value computed as 2.44. Kohn [18], for example, reports the value of Γ_0 as 2.84, while Walsh *et al.* [19] report the value as 2.18. If a value of $\Gamma_0 = 2.65$ were selected within this range, the quality of the fit to shock data (as shown in Figure 1) improves significantly. The same can be done for silver, for which the Grüneisen value was computed [1] as 2.55. Elsewhere, one may find published values from 2.2 in Desloge [20] to 2.47 in Walsh *et al.* [19]. Employing a value of 2.2, as shown in Figure 2, provides an excellent fit to the data.

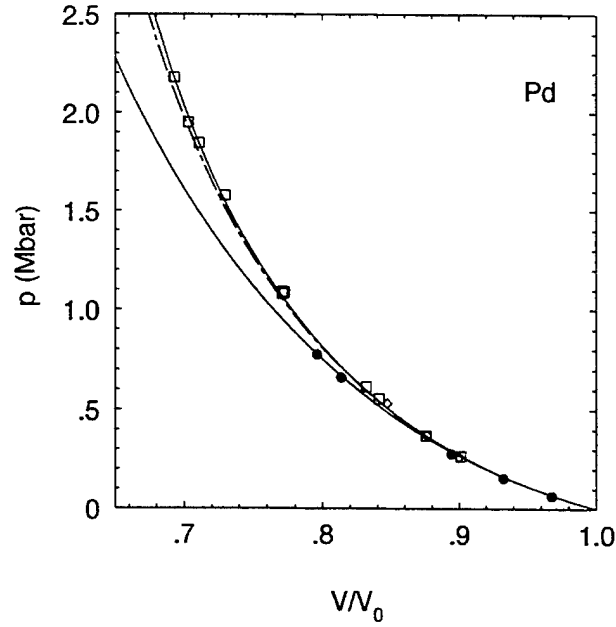


Figure 1. Cold curve (lower) and Hugoniot fits (upper) for palladium. Data and dashed Hugoniot taken from reference [1], with Γ_0 computed as 2.44. Solid line offers improved fit, by selecting Γ_0 in the range of previously published values, at 2.65.

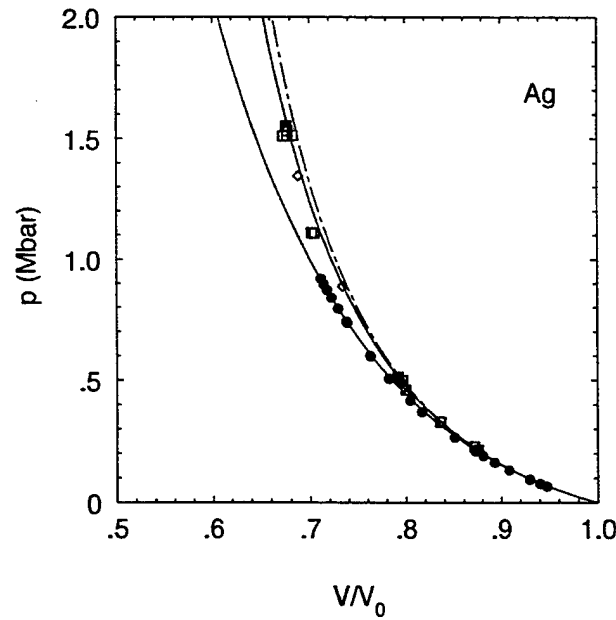


Figure 2. Cold curve (lower) and Hugoniot fits (upper) for silver. Data and dashed Hugoniot taken from reference [1], with Γ_0 computed as 2.55. Solid line offers improved fit, by selecting Γ_0 in the range of previously published values, at 2.2.

4. The Volumetric vs. Vibrational Spring Constant

Segletes [4] examined the relationship between the volumetric spring constant and the vibrational spring constant for an atomic lattice. The use here of the term “constant” is perhaps a misnomer, since these so-called constants are only constant for the case of an idealized linear (*i.e.*, harmonic) spring. In general, these constants will be a function of interatomic distance. The volumetric spring constant is defined as the derivative of the interatomic force, F , with respect to the lattice spacing, λ , or simply $dF/d\lambda$. This force, F , is readily related to the cold (nonthermal) pressure, p_c , by way of the relation, $F = p_c \lambda^2$. The vibrational spring constant, on the other hand, designated $\partial F_{vib}/\partial x$, is that spring constant that, by definition, characterizes the vibrational frequency spectrum of the lattice.

If nothing else were known of the vibrational frequency spectrum, it would be natural to investigate the proportionality of $\partial F_{vib}/\partial x$ and $dF/d\lambda$, since, for a 1-D, simply connected, harmonic lattice, these two quantities would, in fact, be identically equal. The assumption that the characteristic vibrational stiffness of an atomic spring can be made proportional to an associated volumetric stiffness has been pervasive throughout the literature. In addition to proving that Dugdale and MacDonald [6] made this assumption, Segletes [4] showed that this proportionality was assumed explicitly by Brillouin [16] and more recently by Guinea *et al.* [17]. The assumption was also used in an approximate way by Slater [5].

In contrast, Segletes [4] inferred from his equation of state that this proportionality of volumetric and vibrational spring constants does not hold, in general. Whereas the vibrational spring constant, according to vibration theory, should vary as $\partial F_{vib}/\partial x \sim \Theta^2$, Segletes' showed [4], for his idealized quasi-harmonic case, that $dF/d\lambda \sim \Theta [1 + \ln(\Theta/\Theta_0)]$. To infer this distinction from Segletes' equation of state is not to prove the distinction, and since the validity of such an implication would have the effect of overturning a large body of literature that has relied on this assumed proportionality, it would do much to bolster the acceptability of Segletes' equation of state if it could be proven, through other means, that a strict proportionality does not exist between the volumetric and vibrational spring constants. Fortunately, such a proof is easily at hand.

To make this proof, first realize that the interatomic potential for any lattice, expressed as energy vs. lattice spacing, is concave-up over all ranges of compression and into mild ranges of lattice expansion, out to an inflection point. For lattice spacings beyond this inflection point, the energy potential is concave downward out to infinity, whereupon the lattice energy approaches a constant value of zero (if the energy plotted is E_{pot}) or the binding energy (if the energy plotted is E_c). The interatomic force, F , is the (negative of the) derivative of this energy potential curve with respect to atomic lattice spacing, and the volumetric spring constant is the derivative of this force. By the rules of calculus, the interatomic force reaches an extremum, while the volumetric spring constant (the force's derivative) takes on a value of zero at this inflection point in the energy potential. The universal potential of Rose *et al.* [8], for example, shows this effect (see Figure 3). For expansions beyond this inflection point, the volumetric spring constant, $dF/d\lambda$, actually changes sign (which is why the lattice becomes mechanically unstable at that point, as was shown by Guinea *et al.* [17]). As the lattice separation becomes very large, the energy potential becomes flat, and the volumetric spring constant, once again, approaches zero, though this time asymptotically.

The characteristic frequency of the lattice, on the other hand, is a positive, monotonically decreasing function of interatomic distance, which does not reach zero until infinite separation. If the decrease of Θ were not monotonic, eqn (2) tells us that the Grüneisen function would become negative, which does not occur (with the possible exception near phase changes, where behavior is not governed by Grüneisen's assumption in any event). Since vibration theory tells us that $\partial F_{vib}/\partial x$ is proportional to the square of the characteristic lattice frequency, one may logically conclude that the vibrational spring constant is also a monotonically decreasing function of interatomic distance that does not reach zero until infinite separation. Thus, we may logically conclude that a simple proportionality between $\partial F_{vib}/\partial x$ and $dF/d\lambda$ is not possible, since $\partial F_{vib}/\partial x$ is a positive, monotonically decreasing function of interatomic distance, while $dF/d\lambda$ has been shown to be neither monotonic nor exclusively positive.

As to why such a proportionality does not hold, the author believes that the reason lies in the fact that real atomic lattices, unlike most of the idealized lattice models, experience non-nearest neighbor interactions. And though an atom in the lattice is primarily affected by its nearest neighbors, the more distant atoms, too, exert an influence. Let us keep this in mind when we

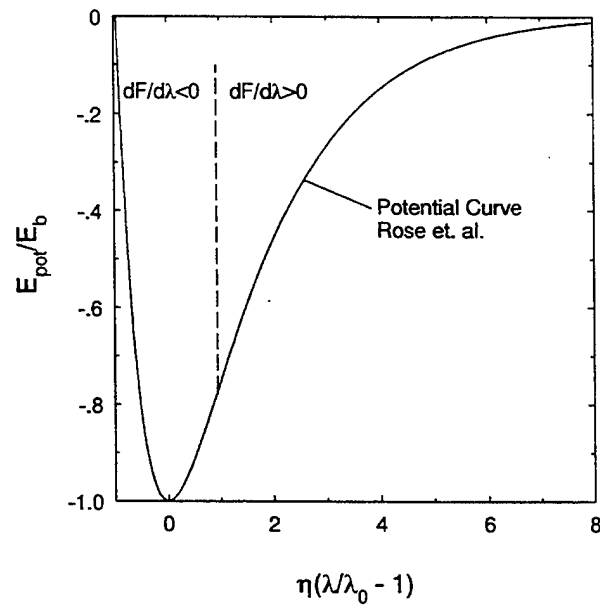


Figure 3. *The energy potential of Rose et al. [8], depicting inflection point and, thus, location of force extremum and volumetric-spring-constant zero.*

imagine the physical processes that go into the makeup of the volumetric and vibrational spring constants. Consider an atom, for simplicity's sake, in a 1-D atomic lattice (a line of atoms), where the point can be made. The volumetric spring constant, $dF/d\lambda$, answers the question of what happens to the interatomic force on this atom when *every atom in the lattice experiences a change in distance* from its nearest neighbor. When measured with respect to our atom of interest, the separation to its nearest neighbors changes by a distance of $d\lambda$, the separation to its second nearest neighbors changes by $2d\lambda$, its third neighbors by $3d\lambda$, and so on.

By comparison, the vibrational spring constant, $\partial F_{vib}/\partial x$, answers the question of what happens to the interatomic force on this atom when *only it moves with respect to a lattice, which is, on average, at a fixed location*. The real problem is more difficult, of course, since all the other atoms, in addition to our atom of interest, are vibrating about their fixed locations as well. Ignoring the vibrations of the rest of the lattice, for purposes of qualitative illustration, the separation distance from our atom of interest measured to any of our atom's neighbors is changed by $\pm dx$ as a result of the vibrational motion of our atom, regardless of whether the neighbor is adjacent to or far removed from our atom of interest. It is quite clear that these two processes are distinct, and a summation of the dF contributions from all the neighbors will likewise be different for these two cases. It is thus equally clear that there is no reason to suspect that measures of those processes, as quantified in the volumetric and vibrational spring constants, respectively, should be functionally proportional. Brillouin [16] realized this situation, as well, and discussed some general properties of the frequency spectrum of a lattice under the conditions of non-nearest neighbor interactions. Most of his rigorous solutions, however, pertain to the simpler case of nearest neighbor interactions only, where one is lured into concluding a proportionality between the volumetric and vibrational spring constants.

5. The Non-Grüneisen Behavior of Aluminum

The one material analyzed by Segletes [1] that seems to cause a great deal of consternation in the readership is aluminum. It is no wonder, since one could argue that aluminum has been unofficially selected as the standard material to which other equations of state are compared. Yet, to fit the form of eqn (1) at moderate and higher pressures, Segletes required a value of Γ_0

equal to 1.20, which is roughly half the value attributed to it at room temperature and zero pressure.

It is only natural, therefore, that one would attribute this discrepancy to Segletes' model [1] and not to the intrinsic behavior of aluminum. Yet, the author has insisted [1–3] and continues to insist that aluminum is not a well-behaved Grüneisen material. This notion is based not upon the discrepancy with the current model, but rather upon earlier equation-of-state stability work [13–15]. It was shown [14] that $d\psi/dV > 0$ is a requirement for the thermodynamic stability of Grüneisen materials. This requirement means that V/Γ must increase with volume—alternately, that Γ can not proportionately change more rapidly than volume. Fits to Γ for aluminum by others (*e.g.*, McKenna and Pastine [21]) show a Γ , which decreases from roughly 2.3 to 1.1 over the relative volume range of $V/V_0 = 1.0$ to 0.82, translating to an average $d\psi/dV$ of -1.73 over that domain. The implication of violating this stability criterion is that, at elevated pressures, $(\partial E/\partial V)_p < 0$. This sort of behavior, in which an energy addition at constant pressure causes material contraction, would generally occur only near a phase change. In aluminum, using this averaged value of $d\psi/dV$, such violations would occur at pressures as low as 200 kbar, where no phase changes are known to occur.

Such a steep gradient in ψ also violates the Hugoniot stability criterion, given as $d\psi/dV > -1$. Under such a violation, it was shown [13] that shocks originating at elevated temperatures, actually can produce postshock pressure values below the preshocked pressure—clearly a nonsensical result. Such behavior does not occur in real materials. Therefore, if one cannot account for this anomalous behavior by the presence of phase changes (and we cannot in the case of aluminum), then the only remaining conclusion is that Γ experiences a temperature dependence at mild elevations of temperature. This is to say that aluminum violates the temperature independence of the Grüneisen assumption. The author has pointed out [1–3] that Grodzka [22] presents data that support this notion. It is indeed ironic that aluminum, the material for which the greatest quantity of shock data likely exists, which is used as a reference material for so many shock experiments, should exhibit behavior that so demonstrably violates the temperature independence of the Grüneisen assumption.

6. Isentropic Transitions

An interesting side note to Segletes' equation of state is that the $\Theta \rightarrow a$ mapping was made on the basis of the complementary solution, eqn (8), to the Grüneisen cold-curve. This same complementary solution, in fact, arises for the solution of any Grüneisen isentrope (whereupon $(dE/dV)_s = -p$), of which the cold curve is but one. If this $\Theta \rightarrow a$ mapping has the validity that the data seem to show, then it would imply that the form of Rose *et al.* [8] is applicable for isentropes, and not isotherms, as was originally asserted by Vinet *et al.* [9, 10].

Such a conclusion, we will see, is also compatible with the lattice specific-heat theories of Einstein and Debye. When considering either theory for the heat capacity of a crystal lattice, an examination of the respective expressions for entropy, S , indicates that entropy for a given lattice remains constant when the quantity Θ/T remains constant, where T is the absolute temperature. Further, both theories indicate that when the quantity Θ/T is held constant, the quantity E_{thrm}/Θ also remains constant, where the subscript “*thrm*” defines the thermal component of the quantity, at a given density.

This condition, which, according to specific heat theory, defines the isentrope, $E_{thrm}/\Theta = \text{constant}$, will be shown to fall out the Grüneisen equation of state as well. To see this, take the Grüneisen equation, eqn (5), with the cold curve as the reference, and express it as

$$P_{thrm}\Psi - E_{thrm} = 0 \quad . \quad (15)$$

Since $(\partial E/\partial V)_s = -p$ and $E'_c = -p_c$, it follows that $(\partial E_{thrm}/\partial V)_s = -p_{thrm}$ as well. So, eqn (15) takes the familiar form of

$$\Psi E'_{thrm} + E_{thrm} = 0 \quad , \quad (16)$$

the solution of which is $E_{thrm} \sim \Theta$. Unlike the cold curve, however, where this solution was merely the complementary solution to the differential equation, the thermal part of the problem is homogeneous to begin with, and, so, this solution defines the particular solution as well for

isentropic transitions. Another way of expressing this solution is $(E_{\text{thrm}}/\Theta)_s = \text{constant}$, which is the same result that arises out of lattice specific-heat theory. And since the characteristic temperature is the independent variable in Segletes' equation of state, this relation provides an extremely convenient method to evaluate isentropic transitions.

7. Conclusions

A variety of subjects were touched upon, in reference to Segletes' equation of state [1–4]. Early feedback that the author has received, both positive and negative, has prompted this report, in an attempt to clarify, explain, and in some cases, prove various facets of the model.

In this report, the basic form employed by Segletes [1], when originally introducing his equation of state, is justified in a more rigorous manner. It is shown how the characteristic temperature is the natural thermodynamic variable that directly relates to both thermal and compressive behavior of a lattice. It is then shown how the theory of differential equations indicates that the complementary solution to the Grüneisen potential, which is proportional to the characteristic temperature, Θ , should correspond to a term in the interatomic lattice potential, in this case, taken as the universal potential of Rose *et al.* [8]. The only mapping between Θ and a term in the potential of Rose *et al.*, which is both unique and meaningful, was the mapping that Segletes chose in his original work [1]. It is shown how the universal potential of Rose *et al.* minus the cubic correction term it contains, corresponds exactly to the potential that results from the idealized form of Segletes' thermal equation of state, known as the quasi-harmonic idealization [4].

The question of the quality of fits achievable by Segletes' equation is then addressed. It is shown that, indeed, excellent fits may be obtained to data, better than those previously reported [1] (which were themselves exceptional, in the author's opinion, though merely adequate, to others). The quality of fit is improved by judiciously selecting the values of the physical parameters, which drive the model, in the range of previously reported (thus plausible) values, rather than just using a single source of data to obtain those parameter values. Such a technique was explicitly avoided in the original work [1], to avoid charges of so-called "parameter tuning."

A proof, employing the mathematical shape of the lattice energy potential, was given to show that a strict proportionality between the volumetric and vibrational spring constants of an atomic lattice cannot exist. The fact that Segletes' model implies a functional uniqueness to these two spring constants had gone counter to a whole body of literature that had derived results based on the assumption of an intrinsic proportionality [4]. By offering the independent proof here that no such proportionality can exist, a possible stumbling block to the acceptance of Segletes' model has been removed. A qualitative explanation was offered as to why, on the atomic level, such a proportionality should not exist.

The behavior of aluminum is, once more, and in greater detail, addressed. The seemingly anomalous behavior is explainable if one accepts the notion that aluminum experiences a temperature dependence of the Grüneisen function at mildly elevated temperatures. It is shown how prior theoretical work supports the notion that aluminum, in fact, violates the temperature independence of the Grüneisen assumption. Such a concept has met with resistance in the readership, perhaps because of aluminum's predominance as a standard material in equation-of-state experiments.

A brief note was made showing how, for an equation of state based on characteristic temperature, isentropic transitions may be easily calculated. Such a method is readily applicable to Segletes' equation of state.

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| 13. ABSTRACT (Maximum 200 words) <p>Clarifications and observations are made regarding Segletes' equation of state, in response to recent comments and criticisms. A theoretical justification for the form of the equation of state is laid out in more precise detail. The model is shown to produce high-quality fits to shock-, as well as cold-compression data, especially when accounting for the experimental variability of the model input parameters. The falseness of the correspondence between the volumetric and vibrational spring constants for an atomic lattice, previously inferred from Segletes' equation of state, is proven by alternate means. The non-Grüneisen behavior of aluminum is addressed, and a brief note is made regarding the nature of isentropic transitions.</p> | | | | |
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